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## Transport Properties of Hydrogen

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The transport properties of hydrogen are calculated for a pressure range from  $10^{-6}$  to  $10^2$  atm and at temperatures up to  $10^6$  °K. The lower temperature limit is taken where the equilibrium gas mixture contains less than 1% molecular hydrogen. The Boltzmann formalism, which assumes binary collisions in the expressions for the transport properties of gas mixtures, is applied. Theoretical values, supported by experimental data whenever possible, are used for the atom-atom, atom-ion, and atom-electron collision cross sections. For Coulomb force interactions between charged particles, the Debye length is used as cutoff distance. The collision integrals for all interactions are computed. Values of viscosity, thermal, and electrical transport coefficients, as well as the thermoelectric coefficients, are presented.

### I. Introduction

DURING recent years the properties of a dissociating and ionizing hydrogen gas have aroused increasing interest, mainly in connection with thermonuclear research, electric propulsion projects, astrophysics, and research in plasma physics. The calculation of transport properties requires knowledge of the interaction potential between the gas particles or their collision cross sections. For molecules, atoms, and ions, these quantities have often been derived by theory and are reasonably verified by experiment. Also, the interparticle forces in a gas consisting solely of charged particles pose no problem. The missing link is constituted by interactions between atoms and electrons in the low energy region. Consequently, the region of dissociation has been under thorough exploration by various investigators.<sup>1-3</sup>

In the present paper, transport properties are calculated in a range where the composition of the gas changes from pure atomic hydrogen to a mixture of neutral particles, ions, and free electrons, and finally to a completely ionized gas.

For the computation of the transport properties, the kinetic theory of gas mixtures is used, which is extensively developed. It has been shown by Grad,<sup>4</sup> Eastlund,<sup>5</sup> and DeVoto<sup>6</sup> that the multicomponent transport theory, as presented by Hirschfelder<sup>7</sup> and Chapman and Cowling,<sup>8</sup> employing only binary collision terms is also valid for the case where charged particles are present. For plasma transport properties, the binary

collision and Fokker-Planck formulations yield identical results.

The composition of the gas was calculated with the chemical equilibrium constants<sup>9-11</sup> using the Newton-Raphson iteration method. Electron capture was found to be unimportant in the pressure range under consideration.<sup>12</sup> Excited states of atomic hydrogen were neglected in this investigation, although their collision cross sections are considerably larger than those of the ground state. The occupation numbers of excited states were calculated with the approximation given by Fermi<sup>13</sup> using a cutoff quantum number of 10. It was found that the occupation number of the first or a higher excited state is in the order of  $10^{-5}$  or less. This result agrees with data given by Marlow<sup>14</sup> and Oppenheim.<sup>15</sup> Thus, excited states of hydrogen contribute a negligible amount to the transport properties in the range of conditions studied here.

### II. Kinetic Theory and Transport Coefficients

#### A. Boltzmann Equation

The calculations of the transport properties are based upon the solution of the Boltzmann integro differential equation, i.e., knowledge of the distribution function taking into account all forces and collisions. In Chapman and Cowling<sup>8</sup> and Hirschfelder<sup>7</sup> detailed derivations can be found; the notation of the latter will be used in this report. The result is that all transport properties can be expressed in terms of collision integrals, which are independent of pressure and functions of temperature only.

The transport properties are expressed in the following equations:

$$j = \sigma E + \alpha \nabla T \quad (1)$$

$$q = -\beta E - k \nabla T \quad (2)$$

$$p = pU - 2\eta S \quad (3)$$

where  $j$  denotes the current,  $q$  the heat flux, and  $p$  the pres-

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sure tensor. The properties  $\sigma$ ,  $\beta$ ,  $\lambda$ , and  $\eta$  are computed from kinetic theory in this paper.

## B. Collision Cross Sections

### 1. Atom-atom interactions

The collision integrals, which enter explicitly the equations of transport properties, have been calculated with good agreement by Grier<sup>1</sup> and Vanderslice.<sup>3</sup> Therefore their data have been adopted for the present calculations of atom-atom interactions.

### 2. Atom-ion interactions

The dominating cross section of collisions between hydrogen atoms and ions is that of symmetrical charge transfer.<sup>16</sup> Close encounters give only a small contribution to the cross section, and their neglect introduces an insignificant error.

The diffusion collision cross section according to Dalgarno<sup>17</sup> simply is

$$Q_{ai}^{(1)} = 2 Q_{ch.tr.} \quad (4)$$

Data given by Fite<sup>18</sup> and Dalgarno<sup>17,19</sup> are shown in Fig. 1. The momentum transfer cross section is taken to be equal to the diffusion cross section. There exist no other data, and the difference is believed small. For this investigation the values of Fite<sup>18</sup> were chosen.

To facilitate later computations, the collision cross sections are plotted as functions of the relative velocity between two colliding particles. The collision cross sections are further chosen to be linear functions in order to execute the required integration without excessive numerical computations. In the range of interest, the deviation of the linear approximation from the calculated case is always less than 3%.

### 3. Atom-electron interactions

The differential cross section as a function of the scattering angle  $I(\chi)$  has been determined analytically by two theories. Burke and Schey<sup>20</sup> used a close-coupling approximation, in which the total wave function is expanded in hydrogen eigenstates, and terms corresponding to the  $1s$ ,  $2s$ , and  $2p$  states are retained. Temkin and Lamkin<sup>21</sup> calculated  $s$ -,  $p$ -, and  $d$ -wave scattering of electrons from atomic hydrogen by the method of polarized orbitals. The results have often been compared<sup>22,23</sup> and were found to agree well except for measurements<sup>24</sup> below 7 eV.

With the available theoretical data at discrete energy levels,<sup>25,20,21</sup> the collision cross sections were determined in an energy range from 1.36 eV to the first excitation threshold by graphical integration according to the equations

$$Q^{(1)} = 2\pi \int_0^\pi I(\chi) \sin\chi (1 - \cos\chi) d\chi \quad (5)$$

$$Q^{(2)} = 2\pi \int_0^\pi I(\chi) \sin\chi (1 - \cos^2\chi) d\chi \quad (6)$$

denoting the diffusion collision cross section and momentum transfer collision cross section, respectively. They are shown in Fig. 2.

Again, the cross sections are computed as a function of the relative velocity of the colliding particles. In the energy range under consideration from 0.1 to 3.5 eV, the cross sections are then approximated by linear functions; their deviation from the computed values is always less than 5%.

### 4. Ion-electron interactions

Theoretical expressions are employed for interactions between charged particles, using the Debye cutoff distance

$$d = [kT/(4\pi n_e e^2)]^{1/2} \quad (7)$$

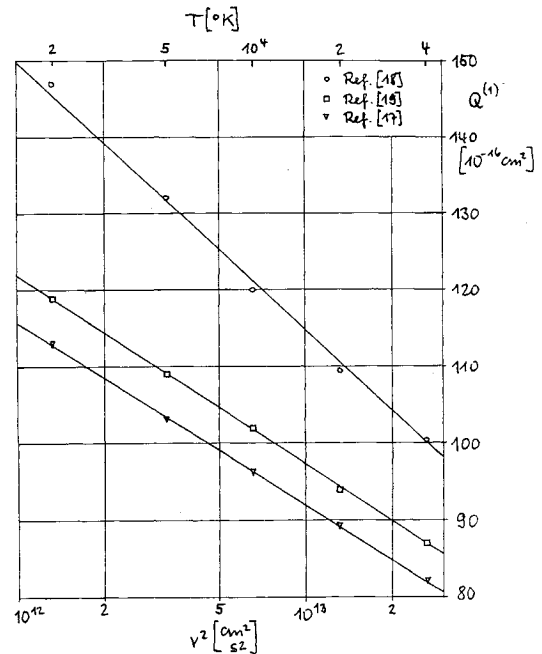


Fig. 1 Hydrogen atom-ion diffusion collision cross section.

## C. Collision Integrals

The collision integrals involve explicitly the dynamics of collisions and are defined by

$$\Omega_{1,2}^{(l,s)} = \left( \frac{kT}{8\mu\pi} \right)^{1/2} \int_0^\infty e^{-x} x^s + {}^1Q^{(1)}(x) dx \quad (8)$$

where  $\mu$  is the reduced mass

$$\mu = m_1 m_2 / (m_1 + m_2) \quad (9)$$

and

$$x = \mu v^2 / 2kT \quad (10)$$

The functions  $Q^{(1)}(x)$  of Eq. (8) have been computed and approximated by linear functions as  $Q^{(1)}(v^2)$  for collisions among the three particles under consideration. Changing the variable of integration from  $x$  to  $v^2$  then produces definite integrals whose solutions are expressions in terms of the gamma function. The values thus calculated are shown in Fig. 3. The collision integrals used for the calculation of transport properties are given in Table 1.

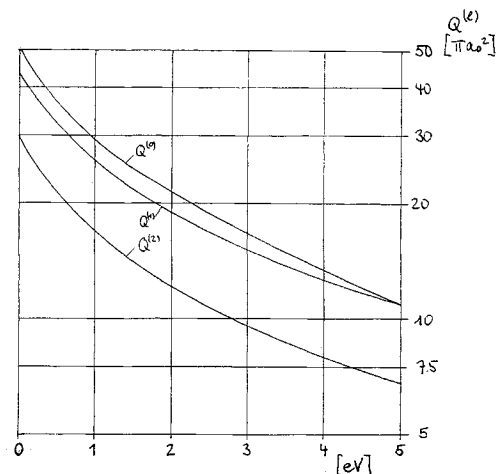


Fig. 2 Hydrogen atom-electron collision cross sections.

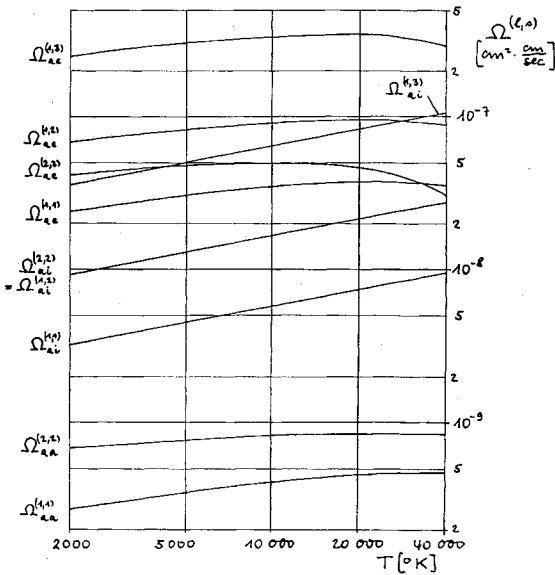


Fig. 3 Collision integrals.

For some much used, simple interaction potentials, the  $\Omega^{(i,j)}$  can be found in the literature. It is currently an active field of research, and collision integrals of more complicated interaction potentials will be published.

Using the collision integral as given in Eq. (8) changes the quantities of Hirschfelder [Ref. 7, Eqs. (8.2-15-8.2-17)] to

$$\begin{aligned} A^* &= \Omega^{(2,2)}/2\Omega^{(1,1)} \\ B^* &= \frac{1}{3}[(5\Omega^{(1,2)} - \Omega^{(1,3)})/\Omega^{(1,1)}] \\ C^* &= \frac{1}{3}(\Omega^{(1,2)}/\Omega^{(1,1)}) \end{aligned}$$

#### D. Viscosity

The first approximation of the coefficient of viscosity  $\eta$  for a gas mixture is given in determinant form by Hirschfelder [Ref. 7, p. 531, Eq. (8.2-25)]. Usually, in solving this determinant, the off-diagonal elements are neglected since their contribution is small, and only the diagonal elements, which contain the viscosity of pure gases, are taken into account. In order to obtain a better approximation, all determinant elements were retained. The equations for viscosity of pure and binary mixtures are taken from Chapman and Cowling (Ref. 8, pp. 162 and 179):

$$\eta_{1,2} = \frac{5}{8}(kT/\Omega_{1,2}^{(2,2)}) \quad (11)$$

and

$$\eta_{1,2} = \frac{5}{8}(\mu_{1,2}/\pi)^{1/2}(k^3/2e^4)(T^{5/2}/A^{(2,2)}) \quad (12)$$

for neutral and charged particles, respectively. The contribution of the pure electron gas is small compared to ions in the ratio of the square root of their reduced masses. In the case of a fully ionized gas, the ion-ion interaction term becomes dominant.

#### E. Thermal Conductivity

The coefficient of thermal conductivity  $\lambda$  for a gas mixture is also expressed in determinant binary collision terms (Ref. 7, p. 1197). The coefficients of pure and binary mixtures are directly proportional to the viscosity

$$\lambda = \frac{5}{2}C_v\eta \quad (13)$$

If the gas is completely ionized, the thermal conductivity can be computed with the Fokker-Planck equation

$$\lambda = 1.90\{[k(kT)^{5/2}]/(m_e^{1/2}e^4\psi)\} \quad (14)$$

where

$$\psi = \ln\{1 + [(4kTd)/e^2]^2\} \quad (15)$$

#### F. Diffusion and Electrical Conductivity

The first approximation of the coefficient of diffusion in a binary mixture  $\mathfrak{D}$  is defined in Ref. 8, pp. 165 and 179 for neutral and charged particles, respectively,

$$\mathfrak{D}_{1,2} = \frac{3}{16}\{kT/[\mu n\Omega_{1,2}^{(1,1)}]\} \quad (16)$$

$$\mathfrak{D}_{1,2} = \frac{3}{16n} \left(\frac{2kT}{\pi\mu}\right)^{1/2} \left(\frac{2kT}{e^2}\right)^2 \cdot \frac{1}{A^{(1,2)}} \quad (17)$$

The second approximations of the coefficients of binary diffusion, calculated with the method given in Ref. 7, p. 606, Eq. (8.A-4), are substituted into the equation of ion-electron diffusion in a ternary mixture, given by Rossini<sup>26</sup>

$$D_{ie} = \mathfrak{D}_{ie} \left[ 1 + \frac{x_a(\mathfrak{D}_{ae} - \mathfrak{D}_{ie})}{x_e\mathfrak{D}_{ai} + x_i\mathfrak{D}_{ae} + x_a\mathfrak{D}_{ie}} \right] \quad (18)$$

The electrical conductivity  $\sigma$  in a three component mixture is given by

$$\sigma = n\xi(e^2/kT)D_{ie} \quad (19)$$

wherein  $\xi$  is the degree of ionization

$$\xi = [x_e/(x_e + x_a)] \quad (20)$$

For the fully ionized gas, the electrical conductivity is given by the Fokker-Planck equation

$$\sigma = 1.97 \{3/[2(2\pi)^{1/2}][(kT)^{3/2}/(e^2\psi m_e^{1/2})]\} \quad (21)$$

#### G. Thermal Diffusion and Thermoelectric Coefficient

The coefficient of thermal diffusion  $D^T$  in a multicomponent mixture can be found in determinant form in Ref. 7, p. 543, Eq. (8.2-53). Considered here is the thermal diffusion of electrons  $D_e^T$ .

The thermoelectric coefficient  $\alpha$  is related to  $D_e^T$

$$\alpha = (e/m_e T)D_e^T \quad (22)$$

The Onsager relation associates the thermoelectric coefficient  $\alpha$  and the electrical conductivity  $\sigma$  by

$$\beta = \alpha T + \frac{5}{2}(kT/e)\sigma \quad (23)$$

thus combining the coefficients of the current and heat flow, Eqs. (1) and (2). The equivalent equation for the fully ionized case is given by

$$\beta = 3.78[(kT)^{5/2}/(m_e^{1/2}e^3\psi)] \quad (24)$$

In the case of gases where temperature gradients and current flow exist, the total heat-flow coefficient is

$$k = \lambda + \beta\alpha/\sigma \quad (25)$$

Table 1 Collision integrals for calculation of transport properties

	Pure gas	Binary mixture	Ternary mixture
Viscosity	$\Omega^{(2,2)}$	$\Omega^{(2,2)}$	$\Omega^{(1,1)}, \Omega^{(2,2)}$
Thermal conductivity	$\Omega^{(2,2)}$	$\Omega^{(2,2)}$	$\Omega^{(1,s)}, s = 1, 3$ $\Omega^{(2,2)}$
Diffusion		$\Omega^{(1,1)}$	$\Omega^{(1,1)}$
Diffusion and approximation		$\Omega^{(1,s)}, s = 1, 3$ $\Omega^{(2,2)}$	$\Omega^{(1,s)}, s = 1, 3$ $\Omega^{(2,2)}$
Thermal diffusion			$\Omega^{(1,s)}, s = 1, 3$ $\Omega^{(2,2)}$

Table 2 Cutoff temperatures

$p$ , atm	$10^{-6}$	$10^{-4}$	$10^{-2}$	$10^0$	$10^2$
$T$ , °K	12,000	16,000	22,000	30,000	40,000
$x_a$	$0.22 \times 10^{-4}$	$0.41 \times 10^{-4}$	$0.65 \times 10^{-4}$	$0.92 \times 10^{-4}$	$0.16 \times 10^{-2}$

or

$$k = 4.58[(kT)^{5/2}]/(mk_e^{1/2}e^4\psi) \quad (26)$$

If stationary solutions are sought with no current flow, a secondary electric field is created which reduces the heat-flow coefficient by the factor  $\beta\alpha/\sigma$ , thus leaving the thermal conductivity  $\lambda$ .

### III. Results

The calculations were performed on the IBM 7094 at the Computer Center of Columbia University. Ternary mixtures were considered up to a cutoff temperature, where  $x_a < 10^{-3}$ . Above that point, the composition was treated as a binary mixture or as a fully ionized gas, as indicated. The cutoff temperatures are shown in Table 2. The computation was carried out stepwise, and the smallest steps were chosen to be 500 °K.

#### Viscosity

The coefficient of viscosity is presented in Fig. 4. At low degrees of ionization, the viscosity is equal to that of pure atomic hydrogen; with increasing ionization it approaches the values of a pure ionic hydrogen gas. The contribution of the electron gas is negligible. The viscosity of binary atom-ion and atom-electron mixtures, entering the determinant as the off-diagonal elements, are several orders of magnitude smaller than the viscosity of pure atoms or ions, a result that is immediately obvious from the collision integrals (Fig. 3), which enter the viscosity according to Eq. (11).

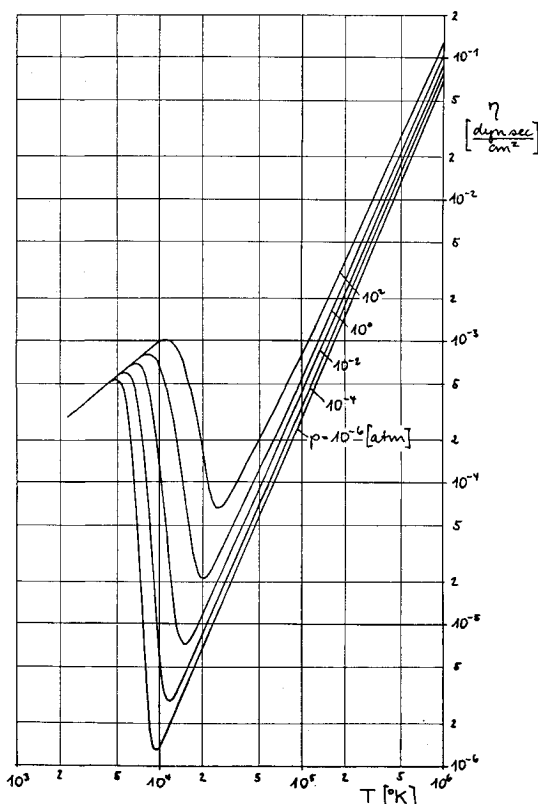


Fig. 4 Viscosity.

Above the cutoff temperature, the results of the binary Boltzmann formalism were compared with the equation

$$\eta_{ii} = \frac{5}{4} \frac{\mu_{ii}^{1/2}(kT)^{5/2}}{\pi^{1/2} e^4 \psi} \quad (27)$$

taken from Marshall.<sup>27</sup> The best agreement is shown at the lowest pressure, within 1%. Values of other pressures differ by 3% or less over the whole temperature range.

#### Thermal Conductivity

Figure 5 shows the zero current coefficient of thermal conductivity  $\lambda$ . Since the conductivity is directly proportional to the viscosity, it is expected that it exhibits similar behavior, which is verified. Atom-atom and electron-electron conductivities are the dominant terms, the latter at high temperatures.

The fully ionized case was calculated using the determinant form from Ref. 7 and the Fokker-Planck equation (14). The results compare very well. For  $p = 10^{-6}$  atm at the cutoff temperature, the binary Boltzmann results are 8% below the numbers obtained with Eq. (14), and they diverge further up to 12% at  $T = 10^6$  °K. Equal or better agreement is shown by the coefficient  $\lambda$  at pressures up to 1 atm. For  $p = 10^2$  atm, however, at the cutoff temperature, the results of the first method are 4% above the calculated values of the Fokker-Planck equation and at the high temperature end 9% below.

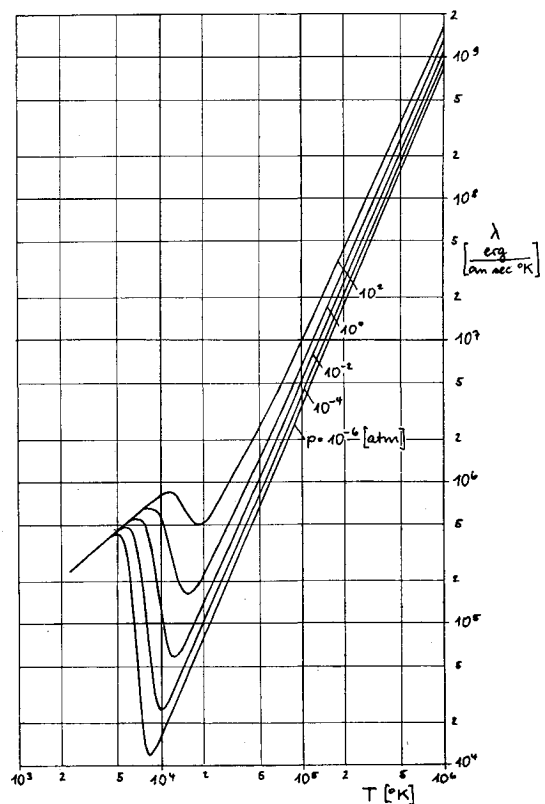


Fig. 5 Thermal conductivity.

### Diffusion and Electrical Conductivity

The binary diffusion coefficients are calculated to the second approximation.  $D_{ai}^{(2)}$  is found to be no more than 18% larger than  $D_{ai}^{(1)}$ , and the values of  $D_{ae}^{(2)}$  are increased by only 1% over  $D_{ae}^{(1)}$  or less, whereas  $D_{ie}^{(2)}$  is approximately 90% larger than  $D_{ie}^{(1)}$ .

The ion-electron coefficient of diffusion in a ternary mixture was computed with Eq. (18). At low temperatures  $D_{ei}$  equals the binary diffusion coefficient  $D_{ae}$ . At higher temperatures it approaches the values of  $D_{ie}$ . The values of  $D_{ai}$  are several orders of magnitude smaller and of no influence on  $D_{ei}$  under the conditions considered here.

The electrical conductivity is shown in Fig. 6 as computed with Eq. (19). For the fully ionized case Eq. (21) was also evaluated. The results of both equations lie within 1.3% or better at the cutoff temperature for all pressures, and between 2.3 to 2.8% at the high temperature end, the better agreement corresponding to the lowest pressure.

### Thermal Diffusion and Thermoelectric Coefficients

The diffusion of electrons due to a temperature gradient yielded very small numbers in the determinants that caused the computer to overflow. Therefore, the values of the thermoelectric coefficients could be calculated only at higher temperatures. The coefficient  $\beta$  is shown in Fig. 7 as computed with Eqs. (23) and (24). At the cutoff temperatures the re-

sults were compared. At this point the values agree within 4% or better, at the highest temperature within 1%.

The total heat-flow coefficient is shown in Fig. 8 for  $p = 10^2$  and  $10^{-6}$  atm. At low temperatures the values of  $\lambda$  are dominating, slowly influenced by the term  $\alpha\beta/\sigma$  of Eq. (25) which becomes noticeable at 3% ionization.

### IV. Discussion

Very limited experimental and theoretical data exist to compare with results obtained in this paper. The assumption that the cross sections are linear functions of the particle velocity introduces a certain error in the integral, which is evaluated from zero to infinity; however, in the range of interest, the deviation is always less than 5%. The integration of a more accurate analytical function would entail considerable additional calculations, a method not justified since the cross sections are not known that precisely.

It is significant that the viscosity of an ionizing hydrogen gas drops by an order of magnitude or more in the pressure range under consideration, whereas the viscosity of dissociating hydrogen virtually remains the same. This sharp decline with increasing ionization follows directly from the much

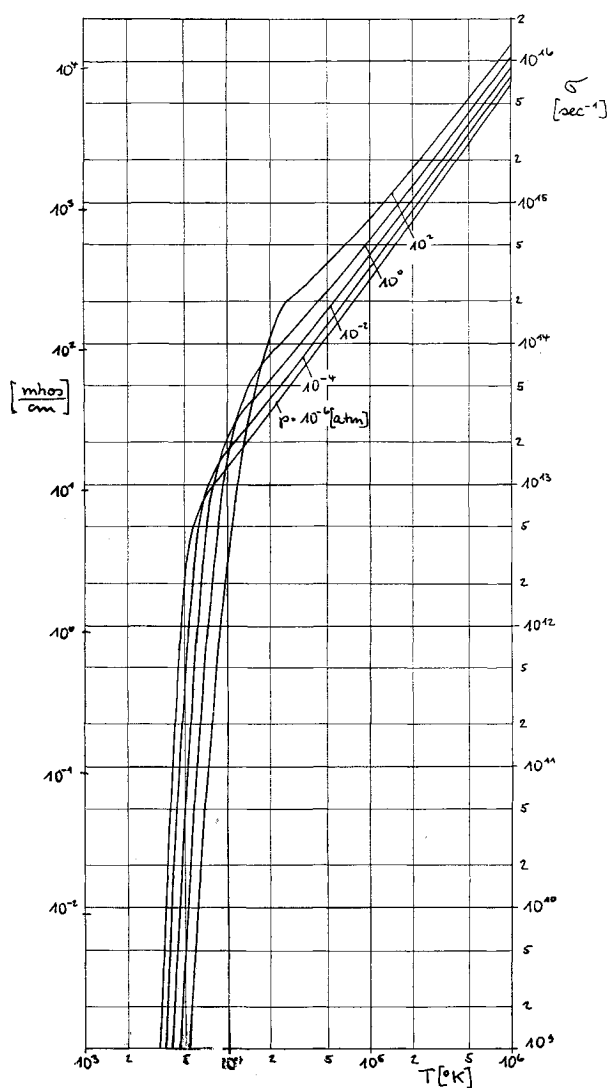


Fig. 6 Electrical conductivity.

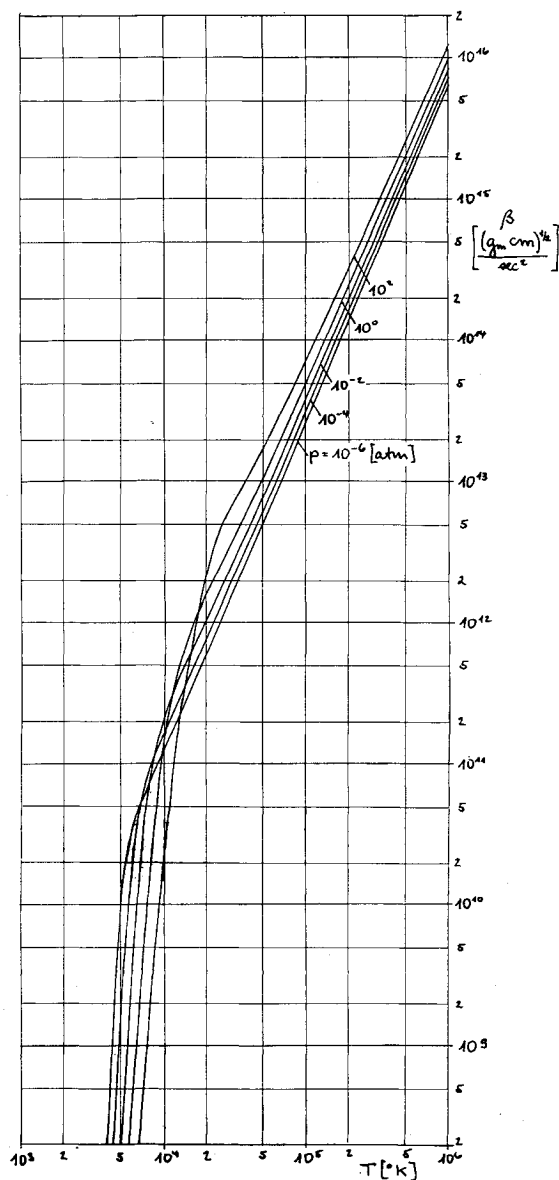


Fig. 7 Thermoelectric coefficient.

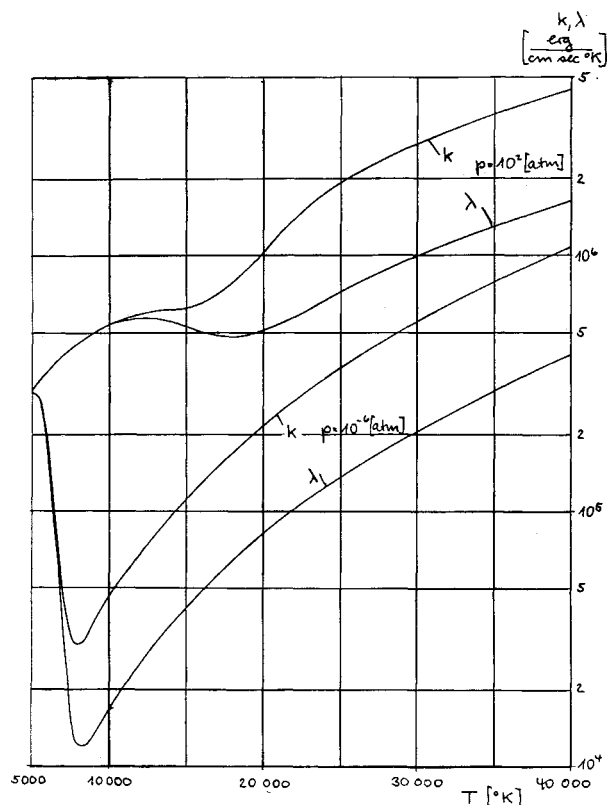


Fig. 8 Heat-flow coefficient  $k$  and thermal conductivity.

larger collision integrals of neutral atoms with charged particles.

The thermal conductivity lies within the expected values, but, of course, does not exhibit the peak at low temperatures which is caused by molecular hydrogen. Curves of the electrical conductivity and the thermoelectric coefficients show the typical crossing over, a behavior familiar from calculations of other elements.

All coefficients calculated with the binary Boltzmann equations agree very well with those obtained from the Fokker-Planck equations, usually within 5%. Better numerical agreement between the first approximation of the binary Boltzmann formalism and the infinite approximation for the fully ionized case cannot be expected.

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